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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

T. IMAI et al

SERIAL NO. 09/840,878

GROUP ART UNIT: 1754

FILED: April 25, 2001

EXAMINER: E. M. JOHNSON

FOR: IRON COMPOUND CATALYST FOR  
INHIBITING GENERATION OF  
DIOXIN AND INCINERATION  
PROCESS OF MUNICIPAL SOLID  
WASTE USING THE SAME

DECLARATION UNDER 37 C.F.R. 1.132

HONORABLE COMMISSIONER OF PATENTS & TRADEMARKS

WASHINGTON, D.C. 20231

SIR:

Now comes Toshiki MATSUI, a citizen of Japan, and a resident of 1-4-605, Miyake 2-chome, Saeki-ku Hiroshima-shi, Hiroshima-ken, Japan, who declares and says that:

1: I graduated from the Department of Ferment Engineering, Faculty of Engineering, Hiroshima University in March 1985.

2. I am currently employed by TODA KOGYO CORPORATION since April, 1985.

3. I am familiar with the work related to U.S. Patent Application, Serial No. 09/840,878, and am a co-inventor of the U.S. Patents: No. 5,993,536 and No. 6,276,287.

4. I have read the Office Action dated January 24, 2003, have understand the Examiner's rejection to the invention claimed in the above application, have studied U.S. Patent No. 5,036,032, and have ascertained the following.

5. Under my control and supervision the following experiments were conducted:

Experiment 1 (Example 1 of U.S. Patent No. 5,036,032)

Silica spheres of nominal 2.2 mm diameter are calcined for 16 hours at 600°C. Surface area measures 80 m<sup>2</sup> /g with a pore volume of 1.0 ml/g. 12.5 g of the spheres are weighed out. 50 g of Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (0.17 mol) are weighed out into a flask and heated to melt the cobalt nitrate salt. Temperature is kept between 85°C and 95°C. No additional water or other solvent is added to the melt. The silica spheres are added to a vacuum filter that contains a 5 to 10 mm layer of 6 mm hollow glass beads. The molten cobalt nitrate solution is poured over the silica spheres with vacuum applied so that the residence time of the molten liquid is approximately 2-4 sec. The spheres are then dried at 120°C. The cobalt loading is 13 wt%. The sample is reduced at 0.2°C/min. from room temperature to 350°C. The dispersion of cobalt is 5.5% corresponding to 170Å diameter crystallites assuming hemispherical particles.

Experiment 2 (Reference experiment of Example 1 of U.S. Patent No. 5,036,032)

There are no examples for preparation of a supported iron catalyst particles in U.S. Patent No. 5,036,032. Therefore, the follow-up test can not be conducted to determine a conversion percentage of carbon monoxide into carbon dioxide of the supported iron catalyst particles. Accordingly, the same procedure as defined in Example 1 of U.S. Patent No. 5,036,032

was conducted except that 69 g of  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  (0.17 mol) is used instead of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , to obtain the supported iron catalyst particles. The dispersion of iron is 5.5% corresponding to 190Å diameter crystallites assuming hemispherical particles.

#### Example 1 of our invention

##### <Production of iron compound catalyst>

126 liters of a 0.4N aqueous sodium hydroxide solution was charged into a reactor maintained under a non-oxidative atmosphere by passing a nitrogen gas therethrough. Next, 12 liters of an aqueous ferrous nitrate solution containing  $\text{Fe}^{2+}$  in an amount of 1.5 mol/liter (the amount of alkali was 1.4 equivalents based on the ferrous iron ( $\text{Fe}^{2+}$ )) was added to the reactor, and then heated to 47°C. After the contents of the reactor were mixed with each other at 47°C for 120 minutes while stirring, air was passed therethrough at flow rate of 70 liters/min for 6.0 hours, thereby producing goethite particles. Thereafter, the obtained goethite particles were successively subjected to filtration, washing with water, drying and pulverization, thereby obtaining aggregates composed of goethite particles.

The thus obtained goethite particles had an average particle size of 0.24  $\mu\text{m}$ , a phosphorus content of 0.002 % by weight, a sulfur content of 0.05 % by weight and a sodium content of 0.08 % by weight, and the conversion percentage of carbon monoxide into carbon dioxide at a temperature of 250°C according to the above specified evaluation method

was 20 %.

Further, the obtained aggregates composed of the goethite particles had a specific surface area of  $2.58 \text{ m}^2/\text{cm}^3$  when measured under a feed pressure of 1 bar in a dry granulometer, and an average particle size ( $D_{50}$ ) of 50 % of a total volume thereof, of  $3.19 \text{ }\mu\text{m}$ .

The properties in the examples were measured by the following methods.

(1) The average particle size of the iron or cobalt particles was expressed by the value measured from an electron micrograph.

(2) The specific surface area of the iron or cobalt particles was expressed by the value measured by a BET method.

(3) The contents of phosphorus and sodium contained in the iron or cobalt particles were expressed by the values measured by an inductively coupled plasma atomic emission spectrometer (SPS-4000 Model, manufactured by Seiko Denshi Kogyo Co., Ltd.).

(4) The content of sulfur contained in the iron or cobalt particles were expressed by the value measured by a Carbon-Sulfur Analyzer (EMIA-2200 Model, manufactured by Horiba Seisakusho Co., Ltd.).

(5) As to catalyst property of the iron or cobalt particles catalyst for inhibiting the generation of dioxin, the catalytic activity was expressed by a conversion percentage of carbon monoxide into carbon dioxide by measuring the concentration of carbon dioxide produced when  $2.8 \times 10^{-4}$  mol of iron oxide particles ( $\alpha\text{-Fe}_2\text{O}_3$ ) obtained by heat-treating the

iron compound catalyst in air at a temperature of 800°C for 15 minutes, were instantaneously contacted with  $6.1 \times 10^{-7}$  mol of carbon monoxide at a temperature of 250°C at an SV of 42,400  $\text{h}^{-1}$  in an inert gas atmosphere using a pulse catalytic reactor.

Here, the "SV" means a space velocity, and is expressed by the value obtained by dividing a flow rate of the reaction gas by a volume of the catalyst. The SV is represented by an inverse number of time ( $\text{h}^{-1}$ ).

The pulse catalytic reactor used comprises a reactor portion and a gas chromatography portion which is constituted by Gas Chromatography GC-16A (manufactured by Shimadzu Seisakusho Co., Ltd.).

The evaluation method used herein was conducted by referring to methods described in the literatures (e.g., R. J. Kobes, et al, "J. Am. Chem. Soc.", 77, 5860(1955) or "Experimental Chemistry II-Reaction and Velocity" edited by Japan Chemistry Institute and published by Maruzen, Tokyo (1993)).

The results are shown in the Table below.

Table

	Properties of iron compound catalyst for inhibiting generation of dioxin		
	Average particle size of iron, cobalt and iron oxide ( $\mu\text{m}$ )	BET specific surface area ( $\text{m}^2/\text{g}$ )	Phosphorus content (wt.%)
Example 1 of our invention	0.25	85	0.002
Experiment 1	0.017	78	N.D.
Experiment 2	0.019	76	N.D.

Table (continued)

	Properties of iron compound catalyst for inhibiting generation of dioxin		
	Sulfur content (wt.%)	Sodium content (wt.%)	Catalyst property (conversion percentage of carbon monoxide into carbon dioxide at $250^\circ\text{C}$ ) (%)
Example 1 of our invention	0.05	0.08	20
Experiment 1	0.01	0.02	7.3
Experiment 2	0.01	0.02	2.1

Remarks

As seen from Experiment 2, the conversion of carbon monoxide into carbon dioxide is 2.1 %, which is more inferior to that of our invention.

As seen from Comparative Examples described in the specification of our invention, on the basis of the lower conversion of carbon monoxide into carbon dioxide than 15 % of our invention, it is presumed that the generation of dioxin can not be sufficiently inhibited.

6. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

7. Further, deponent saith not.

Date: June 9, 2003

Toshiki Matsui  
Toshiki MATSUI